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14. ABSTRACT Optically pumped laser emission is achieved at cryogenic temperatures (<85K) on carbon-implanted nano-patterned silicon-on-insulator. By using ion-implantation and solid-phase-epitaxy for recrystallization, a 30x improvement in the luminescence intensity of silicon is reported. Nano-patterning was achieved through reactive-ion-etching using an anodized aluminum oxide membrane as mask. The results described here lay a solid foundation for the next phase of development aimed at achieving room-temperature lasing in silicon.					
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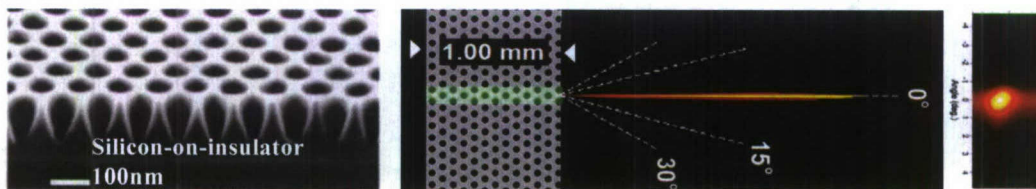
Project Report – 2005-2007

Abstract

Optically pumped laser emission is achieved at cryogenic temperatures ($<85\text{K}$) on carbon-implanted nano-patterned silicon-on-insulator. By using ion-implantation and solid-phase-epitaxy for recrystallization, a 30x improvement in the luminescence intensity of silicon is reported. Nano-patterning was achieved through reactive-ion-etching using an anodized aluminum oxide membrane as mask. The results described here lay a solid foundation for the next phase of development aimed at achieving room-temperature lasing in silicon.

Introduction

Enabled by the ONR support, the research team at Brown University conducted a 3-year study that addressed one of the most interesting questions in semiconductor science and engineering – whether it is possible to make silicon lase. From classical physics, the answer is a clear “No”, for reasons of energy and momentum conservations. But, that conclusion was drawn from our understanding of the silicon crystal structure in its classical bulk phase. The Brown team began their exploration by asking what if that crystalline structure could be altered? In such a case, it is clear that one should not expect the properties of silicon, as determined by the specifics of the crystal structure and symmetries, to remain the same. But what could happen and how to make them happen in ways that silicon lasers become possible? That is a set of fundamental questions they began to probe in this project and one that has broad and profound implications in basic sciences and technologies. Indeed, while silicon electronics has been an unmatched success in modern technologies, silicon photonics is still in development and in need of a laser source. Over the years, many approaches have been explored, from anodized silicon luminescence, to generating direct emissions by quantum-confinement, and to indirect down-conversion of a shorter wavelength laser light via silicon’s nonlinear dielectric responses. One approach that was developed as a result of this project has met with more success than others, and led to the demonstration of laser emission in silicon-on-insulator at cryogenic temperatures ($<85\text{K}$). It is based on nano-patterning of crystalline silicon, as presented in greater details in the section below.

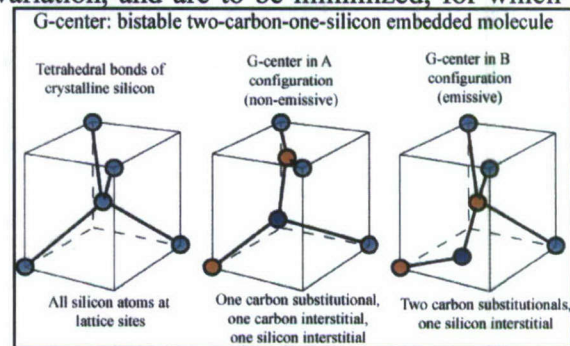


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Findings and results

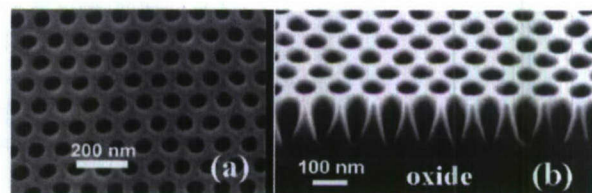
Silicon's inability to emit light and to 'lase' is rooted in the particular atomic arrangement (lattice) of silicon atoms in its crystalline form. As such, the creation of an all-silicon laser or merely an efficient all-silicon light emitter *would necessarily begin at the atomic level*. We took the approach of creating emissive deformation centers (or 'designer defects') in the silicon lattice, an approach that had shown great promise (along with some non-trivial challenges) in our early trials.

These emissive centers exist naturally in silicon. In electronics, they are either detrimental to device performance or are a source of unwanted variation, and are to be minimized, for which purpose they have been extensively investigated since the 60's (e.g. G. W. Watkins, "Defects in irradiated silicon" *Physical Review B* 12, 1975). One example of such centers is called the G-center, which is formed by moving a silicon atom from its normal lattice site and substituting a carbon atom in its place. When the substituted carbon pairs up with a second carbon atom nearby, a local lattice deformation (or, emissive center) is created and an electron captured at the site can then emit light directly. Carbon too exists naturally in all silicon wafers, but is also undesirable in silicon electronics. Its concentration is tolerated if kept below a level of 10^{16} carbon atoms per cm^3 in 'electronic-grade' wafers.



One way to make silicon more optically active is therefore to increase the density of these G-centers, without adversely increasing electrical and optical losses, so as to allow laser action. In considering how to deliberately create these emissive deformation centers, we were fortunate to realise that we had already developed a technique for patterning materials at nanoscale, and that we could apply this technique to silicon, thereby altering its native properties. What we did not expect was that such alteration could be effective enough to make the laser mechanism work under optical pumping, as we found and reported in *Nature Materials* (S. Cloutier et al., *Nature Materials*, 4, 887-891, 2005). More recently, we have built upon this nano-patterning technique and advanced it, by incorporating ion-implantation and solid-phase-epitaxy in the process, and obtained 30x improvement of the emission intensity and an electrically pumped LED (E. Rotem et al. *Appl. Phys. Lett.*, 91, 051127, 2007; E. Rotem et al. *Opt. Express*, 15, 14099, 2007).

The nano-patterning technique we developed is remarkably simple, fast, and low cost. Yet, it proved to be more effective than lithographic alternatives, in scalability, throughput, and feature size, and in preserving the crystallinity of silicon in the unpatterned region. The process uses an etch mask made of a regular array of tiny pores, in a film of anodized aluminum oxide (AAO). In the RIE etching process, this AAO mask, shown in fig. a, is placed directly



on a slice of silicon. The etching through the mask results in a pattern of silicon structures as shown in Fig. b.

For several reasons, using the AAO nano-patterning approach was a fortunate choice that enabled our earlier success in achieving the first silicon laser action under optical pumping (albeit at $<85\text{K}$). The extreme uniformity of the approach helped to keep optical losses low, the large field-size gave us sufficient total optical gain, the small feature-sizes minimized scattering loss, and the hardness of the AAO stood up well against the deep etching process and protected the underlying silicon everywhere except where the nano-pores are to be etched.

The same nano-patterned etching process also created local lattice deformation and strain field in the side-wall region ($\sim 4\text{nm}$ thick) as revealed in TEM and a band-gap narrowing as evidenced in the photocurrent spectroscopy. Both benefited optical emission by facilitating the gathering of electron-hole pairs from the surrounding silicon to the emission centers in the side-wall layer of the etched pore. This itself also benefited from the fact that the nano-pores we create are much closer together than the electron diffusion length within crystalline silicon.

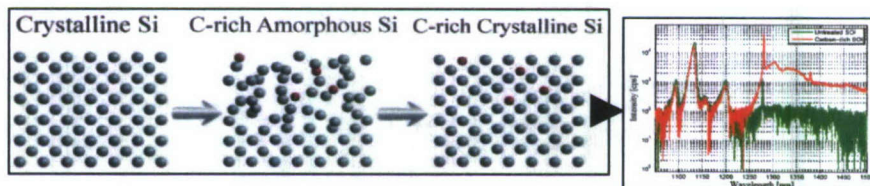
The G-center is just one type of emissive centers. It is better understood and thus explored more in this project. There are other types of emissive centers that may prove to be better at facilitating laser emission at higher temperatures. They too can be made by implantation, that is, by displacement of silicon atoms (without involving foreign atoms) using ion-implantation.

'Ion implantation' through a mask will determine where they do or don't penetrate the silicon. A 400nm thick layer of AAO is found to work well as a mask in this context, as illustrated in the above figures.

We will explore the option to combine patterned implantation with the strain effects that can aid in optical activity, both through direct lattice deformation and indirect selection and stabilization of the desired emissive deformation centers. If needed, biaxial compressive strain induced by epitaxy or strain-field nano-patterning are other tools available to us, and can be used to augment the nanopore patterning and implant-patterning approaches.



Ion-implantation has its own limitation. For example, in the case of carbon implant to create the G-centers, it is limited (to $\sim 10^{17}$ atoms per cm^3) by the solid-solubility of carbon in silicon. To achieve greater optical emission and gain we need to find a way to create more of the emissive centers. To this end, we experimented with the solid-phase-epitaxy (SPE) method, and found that it can work effectively and controllably without compromising the silicon crystallinity. This method takes advantage of the fact that more carbon can be introduced in 'amorphous' silicon. The silicon crystal is pre-amorphized by implanting extra silicon ions, and followed by



carbon ion implantation. The crystal is then heated in a nitrogen or argon atmosphere to induce what is called 'solid phase epitaxy' (SPE): a new layer of emissive silicon crystal, as illustrated in the above figure.

The concentration of the implanted carbon atoms was approximately 10^{19} per cm^3 . However, the concentration of carbon atoms that substitute on silicon sites after re-crystallization may be much lower. Here again, our nano-patterning method came fittingly into the play. It allowed us to create G-centers through the etchant ion bombardment of the silicon lattice, which perturbed the silicon atoms and then allowed the smaller and faster-moving carbon atoms to slip into the vacated silicon positions in the lattice. Our preliminary trial experiments first produced a 30x increase in the luminescence intensity (a spectral comparison of the "before" and "after" nano-patterning is shown in the figure above), and then an electrically pumped LED working at $\sim 30\text{K}$ (limited by internal joule heating).

In the pursuit of the silicon laser, we found ourselves both lucky and vulnerable. Thanks to an innovative lab culture and the lucky combination of right ideas, good people, and relatively simple approaches, we are in the unique position of having been successful in the starting phase of the pursuit – basic concept validation, which laid a good foundation for the next phase of development.

If we succeed, what value can it offer? One answer to these questions is to recall that when the laser itself was first invented, it existed for years as 'a solution looking for a problem', and so we may anticipate that the very ability to create silicon lasers will eventually generate new devices and applications that we have not yet even conceived! There are however, some areas already recognized where true optical and electronic integration could present real benefits, whether in removing the bottleneck of data transmission between the sections of a computer chip, or in mass production of low-cost micro-gadgets that use infra-red light both emitted from and controlled by a silicon chip mounted atop a fiber-scope to diagnose or report medical situations from within a patient.

Publications and Awards

Publications

1. S. Cloutier, R. Guico, and J.M. Xu, "Phonon-localization in periodic uniaxially-nanostructured silicon", Appl. Phys. Lett., **87**, 222104, 2005
2. N. Pavenayotin, M.D. Stewart, J.M. Valles, J.M. Xu, "Spontaneous Formation of Ordered Nano-Crystal Arrays in Films Evaporated onto Nanopore Array Substrates", Appl. Phys. Lett. **87**, 193111 (2005)
3. P.A.Kossyrev, A.Yin, S.G.Cloutier, D.A.Cardimona, D.Huang, P.M.Alsing and J.M.Xu, "Electric field tuning of plasmonic resonances in a gold nanodot-dielectric matrix", Nano Letters, **5**, 1978 (2005)
4. S. Cloutier, P. Kossyrev, J.M. Xu, "Optical gain and stimulated emission in periodic nanopatterned crystalline silicon", Nature Materials, **4**, 887-891, 2005

5. S. Cloutier, C-H Hsu, P. Kosseyrev, and J.M. Xu, "Radiative recombination enhancement in silicon via phonon localization and selection-rule breaking", *Advanced Materials*, 18 (7), 841-844, 2006 (*featured as the cover article in Advanced Materials, April 4, 2006 issue*).
6. Aijun Yin, Marian Tzolv, David Cardimona and Jimmy Xu, "Fabrication of Highly Ordered Anodic Aluminum Oxide Template on Silicon Substrate", *IET Circuits, Devices & Systems*, in press, 2007
7. Jeffrey Shainline, Sylvain G. Cloutier, Chih-Hsun Hsu, Jimmy Xu, "Nano-Engineered Crystalline Silicon for Enhanced Photoluminescence and 1.28 μ m Laser Action – a Study of Mechanisms", *Proc. Of Photonics West*, 2007
8. E. Rotem, J. Shainline, J.M. Xu, "Enhanced photoluminescence from nanopatterned carbon-rich silicon grown by solid-phase epitaxy.", *Appl. Phys. Lett* 91, 051127, 2007
9. Efraim Rotem, Jeffrey M. Shainline and Jimmy M. Xu, "Electroluminescence of nanopatterned silicon with carbon implantation and solid phase epitaxial regrowth" *Optics Express*, Vol. 15, No. 21, 14099-14106, 2007.
10. J. Shainline and Jimmy Xu, (invited) "Silicon as an emissive optical medium" , *Laser & Photon. Rev.* 1, No. 4, 334–348 (2007)
11. J.M. Shainline and J. Xu, (invited) "Directly-pumped silicon lasers", *Optics and Photonics News*, Vol. 19, No. 5, pp. 34-39, 2008

Invited talks and invited conference presentations:

- 1) J.M. Xu, "Periodic Nanometric Superstructures for Photonic Applications", SPIE's Photonics West 22-27 January 2005 in San Jose.
- 2) J.M. Xu, "Explorations of Periodic Nanometric Superstructures in Photonics and Bio-nanoelectronics", Dept. of EECS Seminar, University of Michigan, Feb. 15, 2005.
- 3) J.M. Xu, "Silicon Laser – the impossible is possible", Brown – Oak Ridge Symposium on Imaging and Electron Microscopy, Brown, Dec 13, 2005
- 4) J.M. Xu, "Silicon Laser – impossible possibility", University of North Carolina, Charlotte, NC, Jan. 20, 2006
- 5) Sylvain Cloutier, Chih-Hsun Hsu, J.M. Xu, "Directly pumped all-silicon laser", Optical Society of America Topic Meeting on Silicon Nanophotonics, Uncasville, Connecticut, April 26-28, 2006
- 6) J.M. Xu, "Stimulated emission and emissive structural deformation in nano-patterned silicon", European Materials Research Symposium, Nice, May 29 – June 2, 2006.
- 7) J.M. Xu, "All-silicon Laser – an impossible possibility?" University of New York – Stony Brook, Long Island, NY, June 14, 2006
- 8) Jimmy Xu, "Extending the reach of the mighty silicon technology – to silicon lasers", US-Korea Workshops on Nanomaterials and Nanoelectronics, UCLA, Los Angeles, CA, August 8-9, 2006
- 9) Jimmy Xu, "Directly Pumped Crystalline Silicon Laser - An Impossible Possibility?", the 3rd International Conference on Group IV Photonics, Ottawa, September 13-15, 2006

- 10) Jimmy Xu, "Think big, act small – at the interface between nanoelectronics and biomolecules", Guest Speaker, annual Science Convocation at Lock Haven University of Pennsylvania, Oct. 12th, 2006.
- 11) Jeffrey Shainline, Sylvain G. Cloutier, Chih-Hsun Hsu, Jimmy Xu, "Nano-Engineered Crystalline Silicon for Enhanced Photoluminescence and 1.28 μ m Laser Action – a Study of Mechanisms", Photonics West, January 20-25, 2007
- 12) Jefferey Shainline, Efi Rotem, Jimmy Xu, "Directly Pumped Silicon Lasing", CLEO 2007, Session on "Optical Materials, Fabrication and Characterization", May 7-11, 2007, Baltimore, MD.
- 13) Efi Rotem, Jeffrey Shainline, Jimmy Xu, "Stimulated emission and emission efficiency enhancement in nanopatterned silicon", SPIE Optics East, Sept. 9-12 2007, Boston
- 14) Jimmy Xu, "Silicon Lasers – an anti-physics wish or a real possibility?", Colloquium, Physics Department, Northeastern University, Oct 11, 2007
- 15) Jimmy Xu, "In pursuit of silicon laser", Department of Materials Science and Engineering, National University of Singapore, Jan 22, 2008
- 16) Jimmy Xu, "NanoEngineered Silicon Laser – filling the void of silicon photonics", the 6th "Annual NanoMaterials for Defense Applications Conference", April 21-24, 2008, Washington D.C.

Awards:

1. Guggenheim Fellow, 2005-6, awarded to Jimmy Xu (PI)
2. Named the Charles C. Tillinghast Jr. '32 University Professor, Jimmy Xu (PI)
3. Sigma Xi, 2006, awarded to Sylvain Cloutier (Ph.D student)
4. Brown University Joukowsky Dissertation Award nomination for Sylvain Cloutier
5. Best Engineering Ph.D Thesis Award to Sylvain Cloutier